Accounting for core-core effects in multiconfiguration calculations of isotope shifts

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Isotope shifts

Isotope shift (**IS**)

Key role in extracting *nuclear* properties of an isotope such as its **mean-square nuclear radius** $\langle r^2 \rangle$

Atomic transition k with frequency ν_k

Electronic response of the atom to variations of A & $\langle r^2 \rangle$ given by **only 2** factors :

- **mass-shift** factor M_k
- **field-shift** factor F_k

$\delta\nu_k^{A,A'}$ between any pair of isotopes with A & A' related to $\delta\langle r^2 \rangle^{A,A'}$

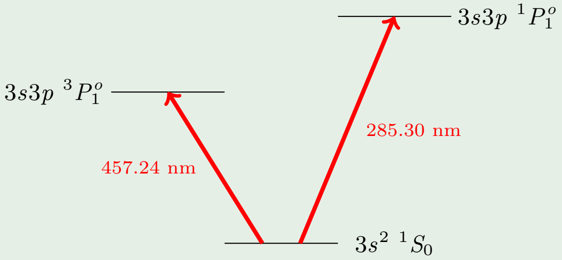
$$\delta\nu_k^{A,A'} \equiv \nu_k^A - \nu_k^{A'} \approx M_k \left(\frac{1}{A} - \frac{1}{A'} \right) + F_k \, \delta\langle r^2 \rangle^{A,A'}$$

Motivation for **Mg I**

Determination of IS in **Mg I**

- Neutral magnesium ($Z = 12$)
  - Ground state [Ne]$3s^2\ ^1S_0$

2 well known transitions from experiments (1979, 2006)



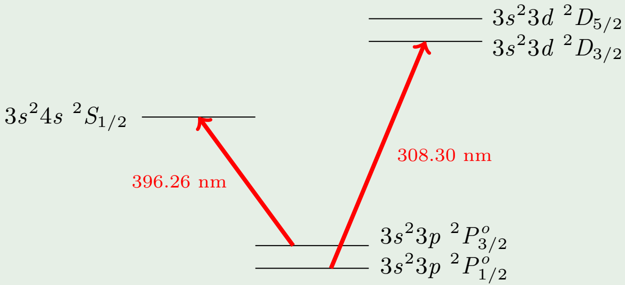
$\Rightarrow$ computation of M_k and F_k factors for these transitions

Motivation for **Al I**

Determination of IS in **Al I**

- Neutral aluminium ($Z = 13$)
  - Ground state [Ne]$3s^2 3p\ ^2P^o_{1/2}$

5 transitions for laser spectroscopy experiments **@ TRIUMF**



$\Rightarrow$ computation of M_k and F_k factors for these transitions

MCDHF Method

Ab initio calculations of IS factors with **Multi-Configuration Dirac-Hartree-Fock (MCDHF)** method (RIS3/GRASP2K)

- Atomic state function (ASF) as linear combination of CSFs :

$$|\Psi(\Pi J M_J)\rangle = \sum_{\nu=1}^{N_{CSFs}} c_\nu \, |\Phi(\gamma_\nu \, \Pi J M_J)\rangle$$

- CSFs constructed from Slater determinants built on Dirac spinors :

$$\phi_{n\kappa m}(\boldsymbol{r}) = \frac{1}{r} \begin{pmatrix} P_{n\kappa}(r)\chi_{\kappa m}(\hat{r}) \\ i\, Q_{n\kappa}(r)\chi_{-\kappa m}(\hat{r}) \end{pmatrix}$$

- **Variational** method : $P_{n\kappa}(r)$, $Q_{n\kappa}(r)$ and c_ν obtained from *self-consistent* procedure that optimizes the energy functional

$$E = \sum_{\mu=1} \sum_{\nu=1} c_\mu c_\nu \langle \Phi(\gamma_\mu \, \Pi J M_J) | H_{DC} | \Phi(\gamma_\nu \, \Pi J M_J) \rangle$$

with $$H_{DC} = \sum_{i=1}^{N} \left(c \, \boldsymbol{\alpha}_i \cdot \boldsymbol{p}_i + (\beta_i - 1)c^2 + V(r_i) \right) + \sum_{i<j}^{N} \frac{1}{r_{ij}}$$

Computational scheme

- M_k estimated from expectation values of one- and two-body recoil Hamiltonian for **a given isotope** + Shabaev relativistic corrections

$$H_{MS} = \frac{1}{2M}\sum_{i,j}^{N}\left(\boldsymbol{p}_i \cdot \boldsymbol{p}_j - \frac{\alpha Z}{r_i}(\boldsymbol{\alpha}_i + \frac{(\boldsymbol{\alpha}_i \cdot \boldsymbol{r}_i)\boldsymbol{r}_i}{r_i^2}) \cdot \boldsymbol{p}_j\right)$$

$\Rightarrow$ 3 contributions : $H_{MS} = H^1_{MS} + H^2_{MS} + H^3_{MS}$

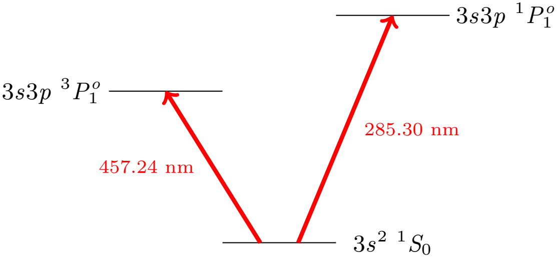
$$H^1_{MS} \Rightarrow \begin{cases} \langle H^1_{NMS}\rangle = \langle \sum_i \boldsymbol{p}_i^2/2M\rangle \\ \langle H^1_{SMS}\rangle = \langle \sum_{i\neq j} \boldsymbol{p}_i \cdot \boldsymbol{p}_j/2M\rangle \end{cases}$$

- F_k estimated from theoretical total electron densities at the origin

$$F_k = \frac{2\pi}{3} Z\, \Delta|\Psi(0)|^2_k$$

where $\Delta|\Psi(0)|^2_k = \Delta\rho^e_k(\mathbf{0}) = \rho^e_u(\mathbf{0}) - \rho^e_l(\mathbf{0})$

Mg I



Active space approach for Mg I

Building the MR

- Common orbital basis for *all* states
- Multi Reference (MR) built from valence-CAS :
  SD from valence orbitals to $n = \{3, 4\}$ shells
- Extract CSFs with mixing coef. $\geq$ threshold **(0.05)**

States	MR	N_{CSFs}
$^1S_0$	$[\mathrm{Ne}]\{3s^2, 3p^2, 3p4p\}$	5
$^{1,3}P^o_1$	$[\mathrm{Ne}]\{3s3p, 3s4p, 3p3d, 3p4s, 3d4p\}$	9

Active space approach for Mg I

(VV + CV) correlations

- SD from valence orbitals + S from $\{2s, 2p\}$ core
- Active orbital set successively extended, including for each new layer one extra l-orbital
- SR-I or MR-I expansions : all CSFs interacting to 1st order with the CSFs defining the SR or the MR

(VV + CV + CC) correlations

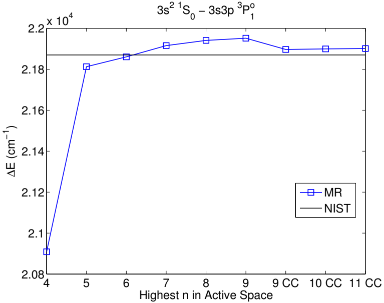
- Convergence reached in (VV + CV) with SD to $n = 9h$ shell
  $\Rightarrow$ **add CC contribution** by CI computation
- SD from ground $1s^2 2s^2 2p^6\ {}^1S_0$ of $\mathbf{Mg}^{2+}$ to $n = \{10, 11\}$ shells
  $\Rightarrow$ **capture only CC contributions**
- CI on each new set of CSFs

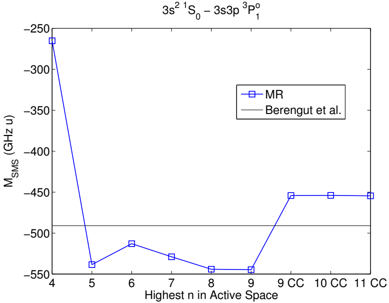
$3s^2\ ^1S_0 \rightarrow 3s3p\ ^3P_1^o$

(VV + CV + CC) correlations

Table 1 : ΔE (cm^{-1}), M_{MS} (GHz u) & F (MHz/fm^2)

Model	ΔE	M_{NMS}	M_{SMS}	M_{MS}	F
$n = 9h$ (VV + CV)	21 952	-286.2	-544.7	-830.9	-77.0
$n = 9h$ CI (VV + CV + CC)	21 897	-356.1	-454.1	-810.2	-76.7
$n = 9h$ + 1 layer CI (VV + CV + CC)	21 900	-356.6	-454.0	-810.6	-76.8
$n = 9h$ + 2 layers CI (VV + CV + CC)	21 901	-356.4	-454.5	-810.9	-76.8
Expt.	21 870	-359.7			



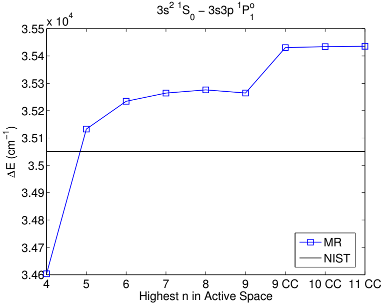


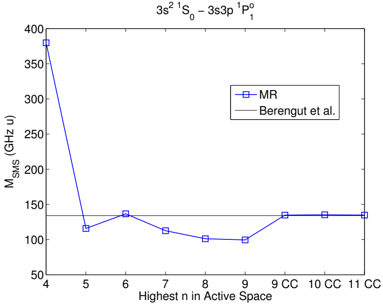
$3s^2\ ^1S_0 \rightarrow 3s3p\ ^1P^o_1$

(VV + CV + CC) correlations

Table 2 : ΔE (cm^{-1}), M_{MS} (GHz u) & F (MHz/fm^2)

Model	ΔE	M_{NMS}	M_{SMS}	M_{MS}	F
$n = 9h$ (VV + CV)	35 265	-506.9	99.5	-407.4	-60.9
$n = 9h$ CI (VV + CV + CC)	35 431	-583.2	134.8	-448.4	-62.0
$n = 9h$ + 1 layer CI (VV + CV + CC)	35 434	-583.7	135.2	-448.5	-62.0
$n = 9h$ + 2 layers CI (VV + CV + CC)	35 435	-583.7	134.8	-448.9	-62.0
Expt.	35 051	-576.4			





Comparison with expt. & Berengut *et al.*

Observed IS between ^{26}Mg & ^{24}Mg

- Comparison with Berengut *et al.* (2005)
  - **field shift neglected** (order of 20-30 MHz) :

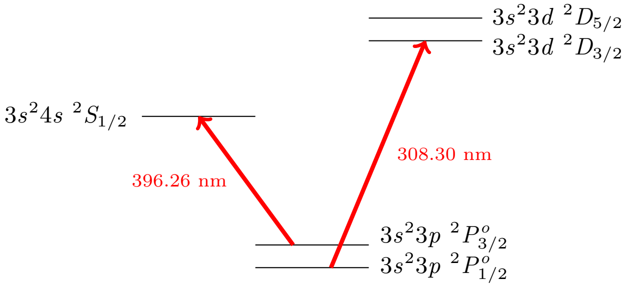
$$\delta\nu_k^{26,24} \equiv \nu_k^{26} - \nu_k^{24} \approx M_k \left(\frac{1}{26} - \frac{1}{24}\right) + \underbrace{F_k\, \delta\langle r^2\rangle^{26,24}}_{\ll MS}$$

Table 3 : IS (MHz) & SMS (MHz) between ^{26}Mg & ^{24}Mg

	IS (MHz)			SMS (MHz)		
Transition	This work	Expt.	Berengut[c]	This work	Expt.	Berengut[c]
$3s^2\,{}^1S_0 - 3s3p\,{}^3P_1^o$	2599	2683(0)[a]	2726	1457	1530[a]	1573
$3s^2\,{}^1S_0 - 3s3p\,{}^1P_1^o$	1439	1412(21)[b]	1420	-432	-436[b]	-428
		1413.8(7.5)[d]				

[a] U. Sterr *et al.*, Appl. Phys. B : Photophys. Laser Chem. **56**, 62 (1993)
[b] L. Hallstadius, Z. Phys. A **291**, 203 (1979)
[c] J. C. Berengut *et al.*, Phys. Rev. A **72**, 044501 (2005)
[d] E. J. Salumbides *et al.*, Mon. Not. R. Astron. Soc. **373**, L41-L44 (2006)

Al I



$3s^2 3d\ ^2D_{5/2}$
$3s^2 3d\ ^2D_{3/2}$
$3s^2 4s\ ^2S_{1/2}$
308.30 nm
396.26 nm
$3s^2 3p\ ^2P^o_{3/2}$
$3s^2 3p\ ^2P^o_{1/2}$

Active space approach for **Al I**

Building the **MR**

- Common orbital basis for *all* states
- Multi Reference (MR) built from valence-CAS :
  SDT from valence orbitals to $n = \{3, 4\}$ shells
- Extract CSFs with mixing coef. $\geq$ threshold **(0.07)**

States	SR	N_{CSFs}	MR	N_{CSFs}
$^2P^o_J$	[Ne]$\mathbf{3s^2 3p}$	2	[Ne]$\{\mathbf{3s^2 3p}, 3s3p3d, 3p^3\}$	9
$^2S_{1/2}$	[Ne]$\mathbf{3s^2 4s}$	1	[Ne]$\{\mathbf{3s^2 4s}, \mathbf{3s3p^2}, 3s3p4p, 3s4s^2, 3p^2 4s\}$	8
^2D_J	[Ne]$\{\mathbf{3s^2 3d}, \mathbf{3s3p^2}\}$	7	[Ne]$\{\mathbf{3s^2 3d}, 3s^2 4d, \mathbf{3s3p^2}, 3s3d^2, 3p^2 3d\}$	14

Active space approach for **Al I**

(VV + CV) correlations

- SD from valence orbitals + S from $\{2s, 2p\}$ core
- Active orbital set successively extended, including for each new layer one extra l-orbital
- SR-I or MR-I expansions : all CSFs interacting to 1st order with the CSFs defining the SR or the MR

(VV + CV + CC) correlations

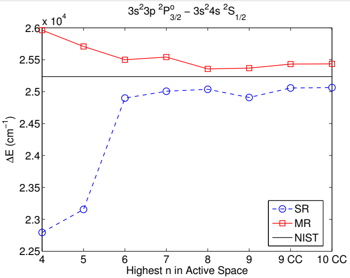
- Convergence reached in (VV + CV) with SD to $n = 9h$ shell
  $\Rightarrow$ **add CC contribution** by CI computation
- SD from ground $1s^2 2s^2 2p^6\ {}^1S_0$ of **Al**$^{3+}$ to $n = \{10, 11\}$ shells
  $\Rightarrow$ **capture only CC contributions**
- CI on each new set of CSFs

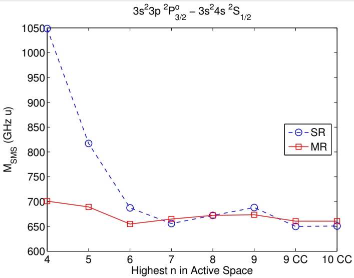
$3s^23p\ ^2P^o_{3/2} \rightarrow 3s^24s\ ^2S_{1/2}$

(VV + CV + CC) correlations

Table 4 : ΔE (cm^{-1}), M_{MS} (GHz u) & F (MHz/fm^2)

Model	ΔE	M_{NMS}	M_{SMS}	M_{MS}	F
$n = 9h$ (VV + CV) SR	24 909	-479.1	688.1	209.0	75.0
$n = 9h$ CI (VV + CV + CC) SR	25 058	-620.8	649.9	29.1	68.9
$n = 9h$ + 1 layer CI (VV + CV + CC) SR	25 064	-629.7	650.9	21.2	68.0
$n = 9h$ (VV + CV) MR	25 369	-418.8	673.5	254.7	79.2
$n = 9h$ CI (VV + CV + CC) MR	25 433	-430.5	660.6	230.1	79.7
$n = 9h$ + 1 layer CI (VV + CV + CC) MR	25 436	-431.2	661.5	230.3	79.6
Expt.	25 236	-415.0			



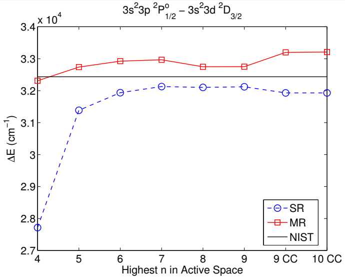


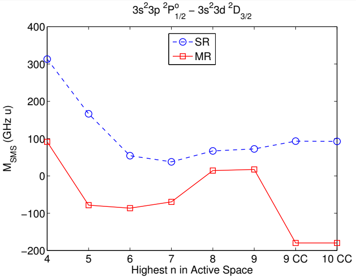
$3s^2 3p\ ^2P^o_{3/2} \rightarrow 3s^2 3d\ ^2D_{3/2}$

(VV + CV + CC) correlations

Table 5 : ΔE (cm^{-1}), M_{MS} (GHz u) & F (MHz/fm^2)

Model	ΔE	M_{NMS}	M_{SMS}	M_{MS}	F
$n = 9h$ (VV + CV) SR	32 122	-541.2	72.4	-468.8	-2.6
$n = 9h$ CI (VV + CV + CC) SR	31 930	-645.5	93.2	-552.3	-1.3
$n = 9h$ + 1 layer CI (VV + CV + CC) SR	31 930	-650.2	92.5	-557.7	-1.4
$n = 9h$ (VV + CV) MR	32 751	-517.0	17.3	-499.7	2.0
$n = 9h$ CI (VV + CV + CC) MR	33 198	-629.7	-179.7	-809.4	-10.5
$n = 9h$ + 1 layer CI (VV + CV + CC) MR	33 208	-630.5	-180.2	-810.7	-10.6
Expt.	32 435	-533.4			





Conclusions & perspectives

Conclusion for **Mg I**

- **CC** contribution **not negligible**
- **Consistent** comparison with nonrelativistic, theory & expt

Conclusion for **Al I**

- $3s^2 3p\ ^2P^o_{3/2} \rightarrow 3s^2 4s\ ^2S_{1/2}$
  - Good agreement for ΔE & F
  - Large difference for M_{MS}
- **Consistent** comparison with nonrelativistic for (VV + CV)

Perspectives for **Al I**

- $3s^2 3p\ ^2P^o_{1/2} \rightarrow 3s^2 3d\ ^2D_{3/2}$
  - **Need for *systematic* procedure to balance the MR**
- Compare with Al I values from RATIP (Prof. S. Fritzsche)
- Compare with experimental data (TRIUMF)

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